

CONF-770646-1

TITLE: SOME RECENT RESULTS ON μ^- TRANSFER IN SYSTEMS WITH BOUND HYDROGEN

AUTHOR(S): J. D. Knight, L. F. Mausner, C. J. Orth, M. E. Schillaci, R. A. Naumann and G. Schmidt

SUBMITTED TO: International Symposium on Meson Chemistry and Mesomolecular Processes in Matter, Dubna, USSR, June 7-10, 1977

By acceptance of this article for publication, the publisher recognizes the Government's (license) rights in any copyright and the Government and its authorized representatives have unrestricted right to reproduce in whole or in part said article under any copyright secured by the publisher.

The Los Alamos Scientific Laboratory requests that the publisher identify this article as work performed under the auspices of the USERDA.

NOTICE
This report was prepared as an account of work sponsored by the United States Government. Neither the United States nor the United States Energy Research and Development Administration, nor any of their employees, nor any of their contractors, subcontractors, or their employees, makes a warranty, express or implied, or assumes any liability or responsibility for the accuracy, completeness or usefulness of any information, apparatus, product, process disclosed, or represents that its use would not infringe privately owned rights.


Los Alamos
scientific laboratory
of the University of California
LOS ALAMOS, NEW MEXICO 87545

An Affirmative Action/Equal Opportunity Employer

MASTER

DISTRIBUTION OF THIS DOCUMENT IS UNLIMITED

SOME RECENT RESULTS ON μ^- TRANSFER IN SYSTEMS WITH BOUND HYDROGEN*

J. D. Knight, L. F. Mausner, C. J. Orth, M. E. Schillaci
Los Alamos Scientific Laboratory, Los Alamos, N.M., U.S.A.

and

R. A. Naumann and G. Schmidt
Princeton University, Princeton, N.J., U.S.A.

For those of us who are attempting to explore and systematize the relationship between chemical structure and muonic and mesic x-ray intensities, one of the more interesting and difficult complications is the effect of hydrogen. We already observe some kind of a correlation, for example, between the apparent radius of an atom (in the pure element or in a compound) and the intensities of the higher members of its muonic Lyman series. The symmetry and polarity of its chemical bonds, and the mode of packing of the crystalline and bulk matter, are probably also significant, although there has been little evidence on these factors. Where the complication enters is that many of the compounds which allow us the greatest variety of chemical structures are hydrogen-containing. These, the compounds of C, N, and O in particular, are also of great practical significance in the biological and industrial areas, and an important goal in our muon chemical effects studies is to determine how and whether the mesic x-ray spectra can be used to obtain information on the chemical constitution of materials of interest.

The hydrogen effect that we refer to was first reported by Budyashov et al.⁽¹⁾, who observed that when negative muons are stopped in the gaseous mixture $H_2 + Ar$, the muonic x-ray spectrum of the Ar displays a marked enhancement of the higher members of the Lyman series ($np \rightarrow 1s$) transitions relative to that of pure Ar. A similar enhancement has been observed in the S and F muonic x-ray spectra from a target of $H_2 + SF_6$.⁽²⁾ Analyses of the $\mu + Z \rightarrow Zu + p$ transfer process involved have identified and characterized the mechanism responsible for population of low angular momentum Zu states that gives rise to the higher member transitions⁽³⁻⁶⁾.

Recently, enhancements of the higher $np \rightarrow 1s$ transitions have been noted also when negative muons are stopped in hydrogen-containing compounds, i.e., structures with bound hydrogen. Comparisons of the muonic x-ray spectra of Li and LiH ⁽⁷⁾ and of C and $(CH_2)_x$ and other compounds containing both C and H^(8,9)

have shown the H-containing form has the higher intensity of the higher series members. Another possible expression of the hydrogen effect has been pointed out⁽⁹⁾ in the muonic x-ray spectra of the alkali halides: the K_β/K_α ratios of the alkali and halide ions in aqueous solution are significantly higher than for the same ions in the crystalline salt. Although the ions in aqueous solution are not "bound" to hydrogens, they are sheathed in water molecules, suggesting the possibility of a contribution from a μ transfer mechanism. Transfer mechanisms in these compound systems would, of course, be expected to be more complex than in the $H_2 + Z$ mixtures. The H is initially bound to the other elements Z, and the abundances of H and Z are approximately equal, so that any μ atoms formed are likely to collide with Z atoms before they have an opportunity to de-excite to their ground state. Indeed, we know from studies of pion capture in hydrogen-containing molecules⁽¹⁰⁾ that very little μ reaches its ground state.

In short, we have some evidence that chemically bound hydrogen has an effect on the $Z\mu$ angular momentum distribution, but the evidence has not yet provided sufficient guidance to enable us to work out the details of the $Z-\mu-p$ encounter.

We describe here the preliminary results of two sets of simple experiments conducted to obtain information on the role of bound hydrogen in possible μ transfer processes by comparisons of muonic Lyman spectra.

In the first experiment we measured the muonic x-ray intensity patterns of Cl in CCl_4 , CH_2Cl_2 , and a $CCl_4 + C_8H_{18}$ mixture. A difference between CCl_4 and CH_2Cl_2 could be ascribed to a change in bonding symmetry or to an intra-molecular μ transfer. A difference between CCl_4 in the pure state and in the mixture with C_8H_{18} could be ascribed to intermolecular μ transfer; we assumed that any change in the Cl intensity pattern, presumably an increase in the intensities of the higher Lyman transitions, would have to have been the result of a contribution from μ transferred from its site of origin in a C_8H_{18} molecule to a neighboring CCl_4 molecule. The results, including also data on the Cl muonic Balmer spectra, are shown in Table IA. The errors listed are the estimated uncertainties in measurement of peak areas and in corrections for muonic x-ray self absorption in the target cell. Since the three liquids were exposed and measured under essentially identical conditions, the other error sources were canceled out from a comparison standpoint. Estimating that $K_\beta/K_\alpha = 0.20$ for a hypothetical Cl spectrum from muon capture in CCl_4 solely

by pp transfer, and assuming initial capture in H, C, and Cl in proportions according to the Z law, we estimate that for the Cl in the $\text{CCl}_4 + \text{C}_8\text{H}_{18}$ mixture K_β/K_α would be about 0.098. As may be seen from the table, the trend of the intensity ratios is roughly in the direction expected for transfer, but taking into account the experimental uncertainties one cannot conclude that intermolecular transfer has been demonstrated. Whatever the mechanism of the intramolecular hydrogen effect, it is shown clearly in the three muonic Lyman spectra for C.

Table IA.

	CCl_4	CH_2Cl_2	$\text{CCl}_4 + 4/9 \text{ C}_8\text{H}_{18}$	$\frac{\text{CCl}_4 \text{ pure}}{\text{CCl}_4 \text{ in mixt.}}$
	Chlorine Spectrum			
K_α	1.0000	1.0000	1.0000	1.00
K_β	$.0905 \pm .0040$	$.0940 \pm .0042$	$.0963 \pm .0046$	$.94 \pm .06$
K_γ	$.0384 \pm .0028$	$.0404 \pm .0032$	$.0413 \pm .0036$	$.93 \pm .10$
K_δ	$.0433 \pm .0031$	$.0441 \pm .0034$	$.0434 \pm .0040$	$1.00 \pm .11$
K_ϵ	$.0300 \pm .0029$	$.0334 \pm .0033$	$.0349 \pm .0037$	$.86 \pm .14$
K_ν	$.0156 \pm .0023$	---	---	---
L_α	$.787 \pm .040$	$.767 \pm .040$	$.737 \pm .040$	$1.07 \pm .08$
L_β	$.115 \pm .005$	$.121 \pm .005$	$.115 \pm .005$	$1.00 \pm .07$
L_γ	$.062 \pm .003$	$.063 \pm .003$	$.067 \pm .003$	$.93 \pm .06$
L_δ	$.035 \pm .003$	$.041 \pm .003$	$.035 \pm .003$	$.99 \pm .12$
L_ϵ	$.013 \pm .004$	$.016 \pm .004$	$.014 \pm .004$	$.92 \pm .39$
	Carbon Spectrum			
K_α	1.000	1.000	1.000	1.00
K_β	$.202 \pm .009$	$.292 \pm .005$	$.342 \pm .003_5$	$.591 \pm .027$
K_γ	$.083 \pm .007$	$.161 \pm .004$	$.186 \pm .002_5$	$.448 \pm .018$
K_δ	---	$.051 \pm .003$	$.0381 \pm .0019$	---
$\left(\frac{\sum \text{Cl } K}{\sum \text{C } K}\right)_{\text{obs}}$	8.6	2.97	1.76	
$\left(\frac{\sum \text{Cl } K}{\sum \text{C } K}\right)_{\text{Z law}}$	11.3	5.67	2.49	

Table IB.			
	C_6F_{14}	$C_6F_{14} + 11.1 C_7H_{16}$	$\frac{C_6F_{14} \text{ pure}}{C_6F_{14} \text{ in mixt.}}$
	Fluorine Spectrum		
K_α	1.000	1.000	1.00
K_β	$.239 \pm .005_4$	$.228 \pm .017$	$1.05 \pm .08$
K_γ	$.211 \pm .005_4$	$.210 \pm .017$	$1.00 \pm .08$
K_δ	$.115 \pm .004_8$	$.125 \pm .014$	$.92 \pm .11$
K_ϵ	$.046 \pm .004_4$	$.072 \pm .012$	$.64 \pm .12$
K_ν	$.028 \pm .006_7$	$(.036 \pm .018)$	
	Carbon Spectrum		
K_α	1.000	1.000	1.00
K_β	$.338 \pm .025$	$.376 \pm .010$	$.90 \pm .07$
K_γ	$.192 \pm .007$	$.222 \pm .010$	$.87 \pm .05$
K_δ	$.050 \pm .009$	$.060 \pm .010$	$.84 \pm .20$
$\left(\frac{\Sigma F K}{\Sigma C K}\right)_{\text{obs}}$	3.80	0.166	
$\left(\frac{\Sigma F K}{\Sigma C K}\right)_{Z \text{ law}}$	3.50	0.185	

The second experiment was conducted like the first, but with F as the test element: the targets chosen for comparison were C_6F_{14} and a $C_6F_{14} + C_7H_{16}$ mixture. It was conjectured that because F has a lower Z than Cl, its normal muon capture might be less dominant and thus capture by $p\mu$ transfer might be more easily detectable than in the CCl_4 case. The results are shown in Table IB. It is apparent that the F spectra show no large effect; only the K_ϵ/K_α ratio is clearly less than unity. However, the trend of ratios in the upper members of the muonic Lyman series is in the direction expected, and we regard this trend as tentative evidence for a $p\mu$ transfer. It is noteworthy that the Lyman intensity pattern for the C in C_6F_{14} differs sharply from that in CCl_4 . This difference is consistent with previous observations that high K_1/K_α in the C spectrum is correlated with short lengths of the bonds joining the C to neighboring atoms.

In the second set of experiments we sought to obtain information on the role of bound hydrogen by comparing the muonic x-ray spectra of the normal and

deutero forms of a compound. Two pairs of targets were used: i) polyethylene, $(CH_2)_x$, one with normal hydrogen isotopic constitution and one with 93% deuterium, and ii) normal water and heavy water (99.8% D). The members of each pair were prepared and exposed under as nearly identical conditions as possible. As with the previous experiments, the muonic x-ray spectra were corrected for target self-absorption and for small non-target backgrounds. To reduce the latter, the usual scintillator telescope used to define muon stops in the target was eliminated and C- and O-containing materials in the vicinity of the target were minimized; the x-ray detector system was gated instead by the accelerator signal.

The relative intensities for the C muonic x-rays from the $(CH_2)_x$ pair and the O muonic x-rays from the H_2O pair are listed in Table II. Also included are the intensities for C in the graphite form. The error limits shown are the uncertainties in the measured peak areas. The numbers in the fourth column are for 100% $(CD_2)_x$; they were computed from the experimental data by assuming the latter to be a linear combination of 7% $(CH_2)_x$ and 93% $(CD_2)_x$ x-ray intensities.

Table II.

Intensity Ratio ($\times 10^3$)	C graphite	$(CH_2)_x$	$(CD_2)_x$	H_2O	D_2O
K_β/K_α	283 ± 3	335 ± 3	323 ± 3	288 ± 3	283 ± 3
K_γ/K_α	147 ± 2	197 ± 2	175 ± 2	258 ± 3	240 ± 3
K_δ/K_α	31 ± 1	57 ± 2	44 ± 2	120 ± 2	103 ± 2
K_ϵ/K_α	4 ± 1	---	---	39 ± 2	27 ± 2

In both the polyethylene and the water samples the isotope effect of the chemically bound hydrogen is clearly apparent in the intensity pattern of the heavier element. We see that the deutero compound has the lower intensity in the higher $np \rightarrow 1s$ transitions, that is, that its Z-cascade contains less contribution from low angular momentum states than does the light hydrogen compound. The comparable magnitude of the effect in the two cases shows that it is relatively insensitive to the details of the bonds to Z; the bonds in

water are all --O--H--O--, whereas in polyethylene there are both --C--C-- and --C--H bonds.

We have not yet succeeded in devising a satisfactory mechanism for the hydrogen isotope effect. Obviously the first requirement is that any proposed model account for the direction of the effect we observe. The earlier observations on the $\text{CCl}_4 - \text{C}_8\text{H}_{18}$ and $\text{C}_6\text{F}_{14} - \text{C}_7\text{H}_{16}$ systems provide some indication that the μ atoms formed can transfer their muons to other heavy atoms in the vicinity, but even if intermolecular transfer were significant it is not evident that the assumed lower velocity of a μ atom would make a noticeable difference in subsequent angular momentum distribution, since the muonic hydrogen atoms are not likely to have had time to de-excite significantly from their original state before encountering a C (or O) atom. A more fruitful approach may lie in considering not the supposed μ migration and transfer but the motions of the original Z-H vibrating subsystem at the time of the μ^- binding encounter, where the differences in the hydrogen isotope mass have an important effect on average Z-H velocity and kinetic energy.

We wish to invite attention to the possibility of another (and unrelated) effect encountered in connection with the foregoing experiments, an effect that we have not yet seen described. In the gross spectra from a number of the simple low-Z targets, including those from background exposures with a Be "stand-in" target, we observe N and sometimes O spectra due presumably to stopping some of the muons in the air in the region viewed by the detector. The interesting feature of these muonic x-ray spectra is that the Lyman series peaks above K_α are missing or only marginally detectable; preliminary estimates give $K_\beta/K_\alpha \approx 0.08$ for both N and O. For a liquid N_2 target, on the other hand, $K_\beta/K_\alpha \approx 0.3$, a "normal" value. These observations, if they are confirmed, would indicate that the muon capture and de-excitation process are density-dependent. Some crude estimates show that the effect is plausible. In liquid N_2 the molecules are in contact, and electrons lost from an $\text{N}\mu$ atom by Auger steps may be replaced from neighboring molecules in times comparable to or less than the de-excitation time. In the gas at atmospheric pressure, on the other hand, the time an atom travels between collisions is of the order of 10^{-9} seconds, and thus on the average its muonic de-excitation cascade will have expelled most of its electrons and gone to completion before collision with another atom. In this case the Auger process would be sooner overtaken by dipole radiation, and the larger Δn steps would bring the atom more rapidly to the $\ell = n - 1$ line, leading only to K_α transitions.

* Work performed under the auspices of the U.S. Energy Research and Development Administration.

1. Yu. G. Budyashov, P. F. Ermolov, V. G. Zinov, A. D. Konin, and A. L. Mukhin, *Yad. Fiz.* 5, 599 (1967) [*Sov. J. Nucl. Phys.* 5 426 (1967)].
2. H. Daniel, H.-J. Pfeiffer, and K. Springer, *Phys. Lett.* 44A, 447 (1973).
3. S. S. Gershtein, *Zh. Eksp. Teor. Fiz.* 43, 706 (1962) [*Sov. Phys.-JETP* 16, 501 (1963)].
4. G. Holzwarth and H.-J. Pfeiffer, *Z. Phys. A* 272, 311 (1975).
5. G. Fiorentini and G. Torelli, preprint, Pisa, April 1975.
6. P. K. Haff, E. Rodrigo, and T. A. Tombrello, *Ann. Phys.* 104, 363 (1977).
7. A. D. Konin, V. N. Pokrovsky, L. I. Ponomarev, H. Schneuwly, V. G. Zinov, and I. A. Yutlandov, *Phys. Lett.* 50A, 57 (1974).
8. H. Daniel, H. Foch, G. Poelz, H. Schmitt, L. Tauscher, G. Backenstoss, and S. Charalambus, *Phys. Lett* 26B, 281 (1968).
9. J. D. Knight, C. J. Orth, M. E. Schillaci, R. A. Naumann, H. Daniel, K. Springer, and H. B. Knowles, *Phys. Rev. A* 13, 43 (1976).
10. A. F. Dunaitsev, V. I. Petrukhin, Yu. D. Prokoshkin, and V. I. Rykalin, *Zh. Eksp. Teor. Fiz.* 42, 1680 (1962) [*Sov. Phys.-JETP* 15, 1167 (1962)]; S. S. Gershtein, V. I. Petrukhin, L. I. Ponomarev, and Yu. D. Prokoshkin, *Usp. Fiz. Nauk* 97, 3 (1969) [*Sov. Phys. Usp.* 12, 1 (1969)]; L. I. Ponomarev, *Ann. Rev. Nucl. Sci.* 23, 395 (1973).